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19. ABSTRACT

The results of a multi-year research effort addressing fundamental scientific issues relevant to the application of laser diagnostic methods as on-line sensors in next-generation propulsion systems are summarized. The overall objective of this research effort was to contribute to the scientific knowledge base necessary to characterize and then extend the capabilities of near-forward scattering (laser-diffraction) particle sizing techniques in terms of application as intelligent sensors capable of on-line, autonomous, and self-diagnosing operation in hostile propulsion system environments. The project scope encompassed three research areas: 1) steering or deflection of the probe laser beam due to refractive index (temperature or concentration) gradients, 2) inverse scattering algorithms, and 3) multiple scattering and measurements in optically thick media. The important technical contributions of this project included: development and demonstration of a concept which allows on-line configuration of optimal detector arrays using transmission-mode spatial light modulators and which can obviate the beam steering problem; derivation of the optimal scaling law for Fraunhofer diffraction particle sizing systems which integrated the optical detector array geometry and the inversion software; systematic formulation and synthesis of the family of integral transform solutions to the inverse Fraunhofer diffraction particle sizing problem and development of a new integral transform; development of a radiation transfer model for near-forward scattering by optically-thick particle media; and development of a general solution and technique for solving the inverse scattering problem for optically-thick dispersions of particles large compared to the wavelength.

RESEARCH ON CERTAIN ASPECTS OF LASER DIFFRACTION PARTICLE SIZE ANALYSIS RELEVANT TO AUTONOMOUS SELF-DIAGNOSING INSTRUMENTATION

Final Report

for

AFOSR Grant 84-0187

for the period

1 October 1984 - 31 May 1990

submitted by

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ABSTRACT

The results of a multi-year research effort addressing fundamental scientific issues relevant to the application of laser diagnostic methods as on-line sensors in next-generation propulsion systems are summarized. The overall objective of this research effort was to contribute to the scientific knowledge base necessary to characterize and then extend the capabilities of near-forward scattering (laser-diffraction) particle sizing techniques in terms of application as intelligent sensors capable of on-line, autonomous, and self-diagnosing operation in hostile propulsion system environments. The project scope encompassed three research areas: 1) steering or deflection of the probe laser beam due to refractive index (temperature or concentration) gradients, 2) inverse scattering algorithms, and 3) multiple scattering and measurements in optically thick media. The important technical contributions of this project included: development and demonstration of a concept which allows on-line configuration of optimal detector arrays using transmission-mode spatial light modulators and which can obviate the beam steering problem; derivation of the optimal scaling law for Fraunhofer diffraction particle sizing systems which integrated the optical detector array geometry and the inversion software; systematic formulation and synthesis of the family of integral transform solutions to the inverse Fraunhofer diffraction particle sizing problem and development of a new integral transform; development of a radiation transfer model for near-forward scattering by optically-thick particle media; and development of a general solution and technique for solving the inverse scattering problem for optically-thick dispersions of particles large compared to the wavelength.

I. INTRODUCTION

Particle and droplet size distributions, being parameters of fundamental importance, should be priority measurement objectives for intelligent sensors in next-generation propulsion systems. However, the potential application of laser scattering systems as optical sensors introduces some severe requirements on the measurement techniques. Optical particle sizing methods can be generally divided into two broad classes, single-particle-counting method which measure the size of individual particles within the relative small optical probe volume, and ensemble techniques which measure the composite or aggregate scattering properties of a particle cloud and use mathematical inversion schemes to determine size distributions from the measured scattering signature. This research is predicated on the premise that autonomous sensors will most certainly come from methods falling in the second category, ensemble techniques. The choice basically involves a speed-resolution tradeoff where ensemble schemes, by virtue of the fact that they analyze the aggregate scattering properties of many particles or droplets at once, provide fewer pieces of more statistically significant information in much less time. Of the various ensemble methods, this research project has focused on the so-called laser- or Fraunhofer-diffraction particle sizing method which uses the near-forward scattering signature to obtain size distribution information. A review of the method prepared during this grant was published by Hirleman [6]. The Fraunhofer diffraction method is generally insensitive to fine details of particle shape and particle refractive index has the potential for on-line calibration and performance verification. This possibility for on-line calibration is quite important for potential applications a autonomous sensors.

The conventional optical system for a laser diffraction particle sizing system which formed the staring point for this research is shown in Fig. 1. A laser beam is collimated to typical several mm diameter and directed through the particle field. A receiving lens is placed after the particle sample to collect the scattered and transmitted (unscattered) light. The transmitted portion of the beam and that light scattered identically in the forward direction (scattering angle $\theta = 0$) is focussed by the receiving lens to a small on-axis spot in the back-focal plane. Light scattered in the near-forward direction (i.e. diffracted) by

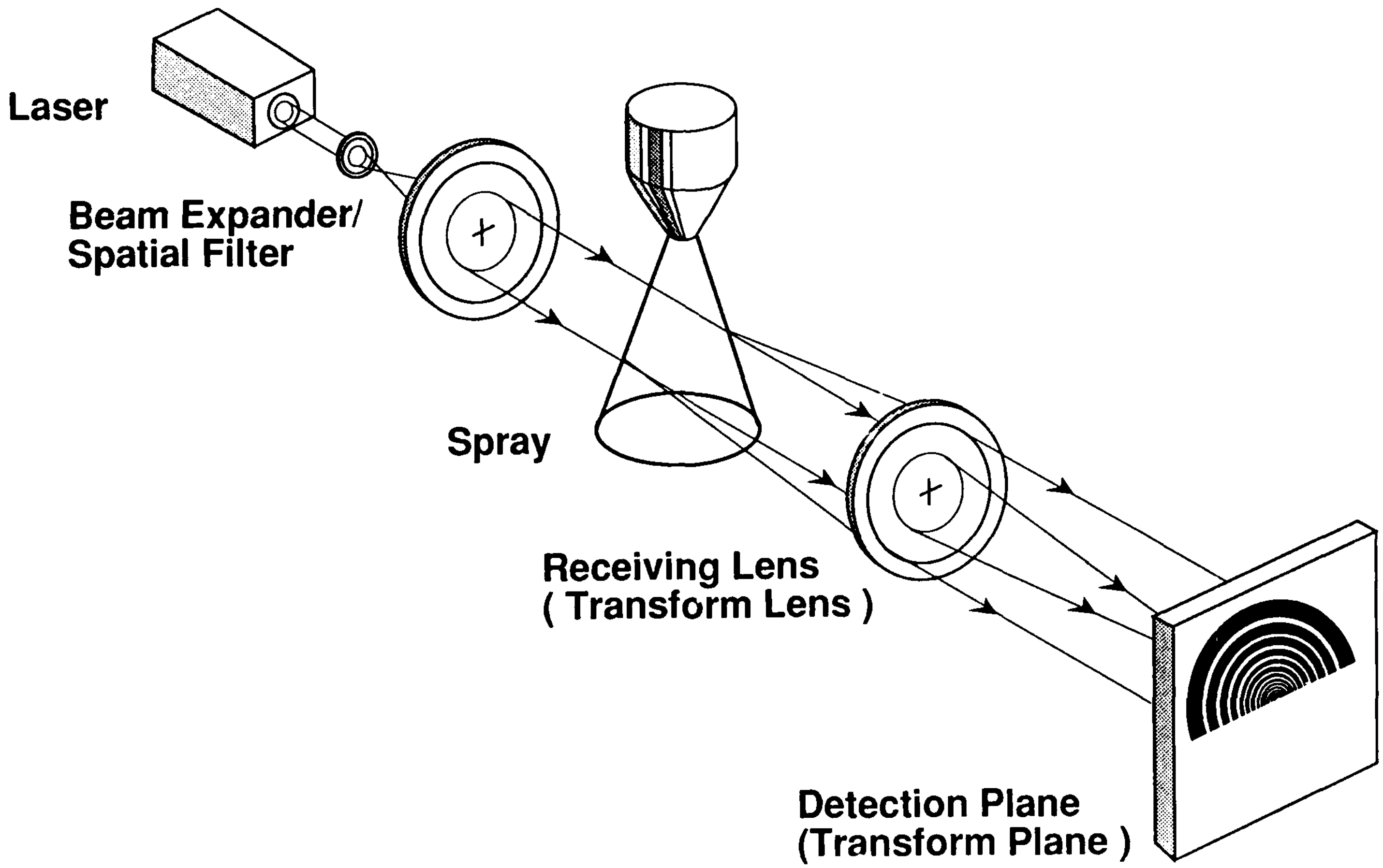

Figure 1. Schematic of Conventional Laser (Fraunhofer) Diffraction Particle Sizing System.

particles along the line-of-sight of the probe laser beam is collected and refracted by the receiving lens to off-axis positions on the back focal plane. (Since the Fourier transform of the E-field distribution in the front focal plane of the lens is formed at the back focal plane, the receiving lens is often called a Fourier transform lens and the back focal plane where the detector array is shown is called the Fourier transform plane). An array of photodiode ring detectors has generally been used at the detection or transform plane in conventional laser diffraction systems. The instrument software attempts to solve the inverse scattering problem for this geometry, i.e. estimate the particle size distribution given a measured forward scattering signature.

In order to apply a system such as that in Figure 1 as an autonomous, on-line sensor to characterize, for example, the properties of a fuel spray in a gas turbine propulsion system, a number of problems must be addressed. First, the question of the robustness and performance of the inverse scattering algorithm must be considered. Clearly, the rate at which measurements of the size distribution can be obtained must be adequate to support on-line control decisions at the frequency required, and this area was identified as deficient when the project began. Secondly, concerning robustness of the inversion algorithms, it is obviously that an instrument and the associated inversion algorithm be intelligent enough to determine some measure of certainty in the results, particularly to recognize faults or bad data. Potential sources of systematic errors in laser diffraction measurements included the effects of multiple scattering and of probe laser beam deflection due to thermal or species concentration gradients in the propulsion environment. These areas too were identified as presenting critical impediments to future sensor implementations and fell within the scope of this research effort.

In the sections below, the results of the project are discussed in terms of the research objectives as stated in the original and continuation proposals. This final report is intended as an overview and synthesis of the research results as opposed to an exhaustive restatement of what has appeared in previous progress reports, annual reports, and published technical papers. The next sections of this report provide a quasi-chronological summary of the research as it evolved, organized in terms of the research objectives from the original proposal. Publications resulting from this work are incorporated by way of references from the technical section. Finally, a summary of personnel involved in the project is presented.

II. LASER BEAM DEFLECTION/STEERING

A problem which has already limited the usefulness of laser diffraction particle sizing instruments in propulsion systems research is that of beam steering or deflection by refractive index gradients in the optical path. These gradients are due to spatially nonuniform temperature and/or species concentrations and at relatively large scales have the same effect as an array of randomly dispersed and time-varying lenses which cause the probe beam to move around at the detection plane. This has been a severe problem since information on the largest droplets in typical sprays is concentrated at small scattering angles which are of the same order as the beam deflection angles encountered.

The objective of this component of the research was to investigate concepts which could enable laser diffraction particle sizing sensors to operate under conditions where beam deflection is significant. A minimal requirement is for on-line detection of beam steering so that an intelligent instrument would at least know that the data taken under these conditions is suspect. A more important objective is to develop strategies to permit a diffraction system to autonomously perform real-time correction for beam steering.

The system we developed and demonstrated for this problem is shown in Fig. 2 which initially appeared in our Annual Report to AFOSR covering the period 1 October 1984 - 1

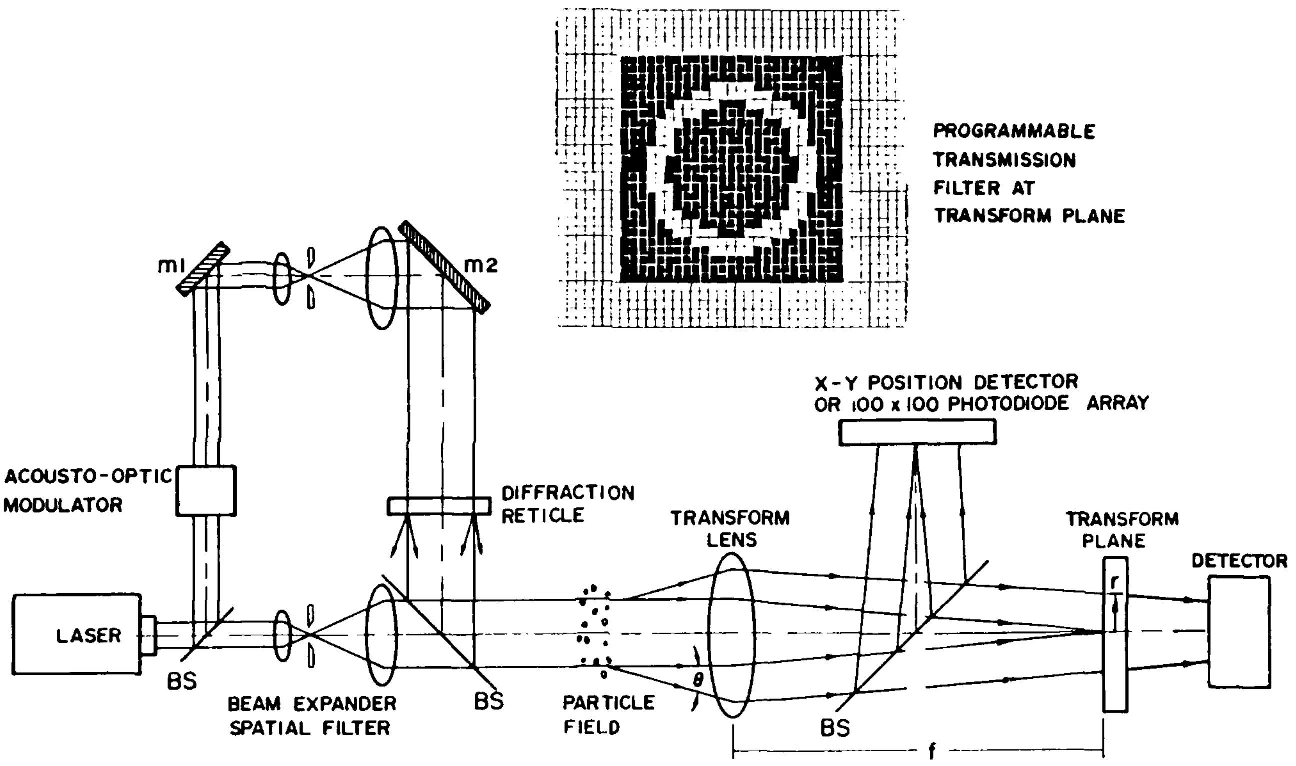

Figure 2. Schematic of proposed laser diffraction particle sizing system which incorporates on-line detector array configuration and solves the beam steering problem. Both the modulated diffraction signature from the calibration reticle and the portion of the probe laser beam scattered by the particles are collected by the transform lens. A sequence of ring-shaped apertures (see inset) are created in the transmission filter (light valve array) array concentric with the instantaneous beam center using feedback from the x-y position detector which monitors beam deflection. Light transmitted through the light valve array is detected by the field detector.

October 1985. The x-y position detector or a rectangular photodiode array is used to track in real-time the position of the center of the probe laser beam (and the associated diffraction pattern). This information of the instantaneous position of the laser beam can be used in two ways.

First, for fixed detector arrays as in the case of the photodiode ring detectors used in Malvern and other commercial instruments, the position sensor output could be used as a control signal to ensure that the detectors would be sampled *only when the probe beam is centered in the ring array at the detector plane.* The fraction of time for which the beam centering was acceptable would depend on the flow conditions, but at those discrete times when a measurement was possible the data would be valid. This conditional sampling approach has been subsequently demonstrated by the Purdue University group (see paper by B. H. Miles, G. B. King, and P. E. Sojka, pp. 225-230, in *Optical Methods in Flow and Particle Diagnostics*, Vol. 63, Laser Institute of America, 1988).

A second approach which we studied and that can potentially provide continuous size distribution data involves the use of programmable detector arrays to "follow" the instantaneous beam center around the detection plane. The approach could also be achieved using the system in Fig. 2, where the probe beam center measured with the x-y position detector would be used as the origin of a dynamically configurable detector array. In our research we used a 48x48 array of light valves (transmission mode spatial light modulator) as reported by Hirleman and Dellenback [10,11,15]. An algorithm to determine the beam center at the detection plane by opening a sequence of groups of the light valve pixels was developed and demonstrated. After the beam center was located, a sequence of concentric ring detectors were opened in the optical shutter array centered about this origin [10,11,15]. While the system we demonstrated was relatively slow (Hz) and of low resolution (48 x 48 pixels), feasibility of the concept was successfully demonstrated. It appears that with some reasonable improvements in spatial light modulator technology (resolution, contrast ratio, efficiency, and speed) an instrument based on this concept could be useful as a propulsion system sensor.

III. INVERSE SCATTERING ALGORITHMS

The first research area targeted in this work is that of inverse scattering algorithms. Particle sizing using laser (Fraunhofer) diffraction is a classical inverse scattering problem. The governing equation for the Fraunhofer diffraction particle sizing problem is a nonhomogeneous Fredholm integral equation of the first kind which has been formulated by Hirleman [9,12] as:

$$i_a(\theta) = I_{inc} / k^2 \int_0^\infty J_1{}^2(\alpha\theta) / (\theta^{2-a}\alpha^{b-2})\, n_b(\alpha)\, d\alpha \tag{1}$$

Equation (1) is valid in the far-field and predicts the near-forward (small-angle) light-scattering signature of particles significantly larger than the wavelength with refractive indices significantly different than the surrounding medium. In Eq. (1), $i_a(\theta) \equiv i(\theta)\theta^a$ and $i(\theta)$ is the intensity (W/sr) diffracted at scattering angle θ measured from the incident beam propagation direction, I_{inc} is the irradiance (W/m^2) assumed to be uniformly incident on all particles sampled by the optical beam, α is the size parameter (defined as $\pi D/\lambda$), D is the particle diameter, λ is the wavelength of the incident beam, k is the wavenumber (defined as $2\pi/\lambda$), J_1 is the Bessel function of the first kind and of order one, θ is the scattering angle measured from the incident beam propagation direction, and $n_b(\alpha) \equiv n(\alpha)\alpha^b$ where $n(\alpha)d\alpha$ is the number of particles in the laser beam with sizes between α and $\alpha + d\alpha$ (i.e. $n(\alpha)$ is an

unnormalized probability density function). In Eq. (1) a small angle approximation of $\sin\theta \approx \theta$ was made, and vignetting and multiple scattering were assumed negligible.

To extend Eq. (1) and account for cases where Fraunhofer diffraction theory is not adequate we can write:

$$i_a(\theta) = I_{inc} / k^2 \int_0^\infty k_{ab}(\alpha,\theta)\, n_b(\alpha)\, d\alpha \tag{2}$$

where $k_{ab}(\alpha,\theta)$ is the scattering function or kernel of the integral equation and for the diffraction case we have:

$$k_{ab}(\alpha,\theta) = J_1^2(\alpha\theta) / (\theta^{2-a}\alpha^{b-2}) \tag{3}$$

In this research, we have considered cases where Eq. (3) is valid and the more general situation where the Fraunhofer diffraction approximation is not adequate and the full Lorenz-Mie theory solution or a better approximation must be used. For the general case of arbitrary particle size and refractive index a kernel function $k_{ab}(\alpha,\theta)$ based on the full Lorenz-Mie scattering functions would be required in place of Eq. (3) for use in Eq. (2).

Solutions Involving the Linear System Produced by Quadrature

The essence of the Fraunhofer diffraction particle sizing problem is to detect and analyze the diffraction signature, $i_a(\theta)$, and mathematically invert Eq. (2) to determine the particle size distribution, $n_b(\alpha)$. One general approach to all inverse problems involves solving the linear system resulting from numerical quadrature. Specifically, Eq. (2) is discretized into a finite number of size classes and detector elements, whereby the scattering measurement is a vector with n elements. This measured scattering signature vector is denoted as **S** such that the ith element of **S** corresponds to the measured scattering contribution on the ith detector. Now the resulting linear system has n equations (supported by the n measurements in **S**) and therefore may be solved to obtain n unknowns, i.e. the particle populations in n discrete size classes. If the vector **N** contains as the jth element the quantity of particles in the jth discrete size class, then the linear system is written:

$$\mathbf{S} = \mathbf{K} \cdot \mathbf{N} \tag{4}$$

Now the elements of K depend on the values of the parameters a and b from Eq. (1) and on the assumption of the scattering model, as in Eq. (3). The most straightforward approach to solve Eq. (4) for the particle size distribution contained in **N** is to invert the matrix **K**:

$$\mathbf{N} = \mathbf{K}^{-1} \cdot \mathbf{S} \tag{5}$$

where $\mathbf{K}^{-1}$ is the inverse of **K**. Unfortunately, the matrices **K** determined by Eq. (2) are generally very ill-conditioned, and can not be inverted directly. Hirleman [6,12] has discussed this problem in some detail, particularly from the point-of-view that the optical sensor designer should consider the condition of **K** in the design of the laser diffraction sensor. One measure of the expected performance of an inversion such as in Eq. (5) is the condition number of **K** and Hirleman [12] has studied cond(**K**) as a function of the diffraction sensor design parameters and proposed the optimal scaling laws for such a sensor. These optimal scaling laws, one contribution of this research project, encompass the detector geometry number, mean scattering angle, and geometry) and the size classes (mean size and width) and will most certainly be used in the design of future laser diffraction systems.

Another advantage of examining the condition number of **K** is the ability to consider the effects of noise on the inversion performance. Since the condition number is a measure of the amplification of noise by the mathematical inversion process, when the noise/signal ratio becomes of the order of 1/cond(**K**) then the inversion will become unstable. Since cond(**K**) can be improved by reducing the order of the linear system in Eq. (4), i.e. by reducing the number of degrees of freedom in the solution vector **N**, then an intelligent system based on the results of Hirleman [12] will be able to diagnose on-line (in real time) the measurement context and determine the optimal number of detectors in addition to their optimal positions and geometries. This, however, requires programmable detector arrays and provides another reason for our research on methods for creating programmable arrays. Our research considered the use of transmission mode spatial light modulators to provide the capability to reconfigure on-line these diffraction detector geometries. Figure 2 shows a schematic of an optical system which we have developed and studied in detail and which might form the basis for next-generation intelligent diffraction sensors. The papers of Hirleman and Dellenback [10,11,15] provide a detailed description of these devices and our research on their use in laser diffraction particle sizing systems.

Integral Transform Solutions

As stated earlier, inverse scattering problems can be solved by direct integral transform solutions in addition to the indirect methods based on numerical quadrature of the governing integral equation as just discussed. Because direct integral transform solutions potentially could provide must faster time response in a laser diffraction sensor since the iterations are avoided, we also considered integral transform solutions to the governing Eq. (1).

In the past three decades, five integral transform solutions to Eq. (1) were proposed which we considered in detail during this work. Unfortunately, the individual formulations used different notation and the interrelationships remained very unclear until the series of publications resulting from this project [3,4,8,16,18]. Further, no definitive, quantitative work comparing the performance of the various methods was available previously. The overall goal of our research was to provide an objective evaluation of integral transform methods in terms of potential performance in autonomous sensors. In that spirit each method was derived and the entire family of transforms placed in context in a systematic framework. Three of these previously existed in the literature, one had errors in derivation which we corrected, and the fifth was newly developed as part of this work.

Equation (1) is a classical Fredholm integral equation of the first kind which is solved to determine the particle size distribution function $n_b(\alpha)$ subject to the diffraction theory approximation. The numerical solution of a Fredholm integral equation of the first kind is an ill-posed problem. The integral transform solutions are closed-form solutions to Eq. (1) which make them potentially much faster than the inversion or iterative steps that are required in the numerical quadrature methods. More than one integral transform solution exists since a and b are essentially instrument parameters, and different solution expressions are obtained for different instrument configurations. Also, simplification of the expression for $k_{ab}(\alpha,\theta)$, for example by using asymptotic expansions, can also provide different direct solutions.

In order to facilitate representation of the family of integral transforms considered here we developed a general form for the integral transform solution equation:

$$n_b(\alpha) = -k^2/I_{inc} \int_0^{\infty} h_i(\alpha\theta)\ d/d\theta[e_i(\theta)]\, d\theta \tag{6}$$

where the subscript i indicates the ith form for the integral transform solution, $h_i(\alpha\theta)$ is the ith kernel function, $e_i(\theta)$ is a function derived from the experimental (scattering) measurements, and the solution provides an estimate related to the bth moment of the unnormalized particle size distribution on a number basis. Now the first solution presented we have designated the Chin-Shifrin solution [18] and can be written:

$$n_2(\alpha) = -(k^2/I_{inc}) \int_0^\infty [2\pi J_1(\alpha\theta) Y_1(\alpha\theta)\alpha\theta] \; d/d\theta[i_3(\theta)] \, d\theta \tag{7}$$

from which, based on a comparison with Eq. (6), we can determine:

$$h_{CS}(x) = -2\pi J_1(x) Y_1(x) x \tag{8}$$

$$d/d\theta \, [e_{CS}(\theta)] = d/d\theta \, [i_3(\theta)] \tag{9}$$

Now another solution to Eq. (1) can be obtained through an integration by parts of Eq. (6) or (7) such that new solution functions h_i and e_i are related to the previous ones by integration (in the case of e_i) and differentiation (in the case of h_i).

In a series of publications resulting from this research grant we have derived a new member of the integral transform family [8,18], formulated the entire family of transforms using a consistent set of variables [18], developed eleven statistical criteria to quantify the accuracy and stability of each solution technique [3,8], and finally compared the performance of the various techniques on a carefully-selected set of benchmark test cases [4,8,18]. Our research has shown that the original (Chin-Shifrin) integral transform had the best overall performance for the range of numerical experiments we completed. This method was able to reconstruct trimodal size distributions using scattering data for a simulated diffraction instrument configuration and noise levels which should be attainable in sensor systems.

This conclusion received some further independent substantiation from the study of Hirleman [14] of analytic eigenfunction expansion solutions to Eq. (1). These solutions exist only when the kernel function $k_{ab}(\alpha,\theta)$ is a function of only the product of the two variables, i.e. $k = k_{ab}(\alpha\theta)$. With this requirement we see from Eq. (3) that:

$$2 - a = b - 2 \tag{10}$$

or:

$$a + b = 4 \tag{11}$$

and we have lost one degree of freedom in scaling the problem as a and b are no longer independent. Equation (11) is the same condition for which the linear system produced by numerical quadrature of Eq. (1) takes the Toeplitz form as discussed by Hirleman [12]. If we define a new independent scaling parameter δ as:

$$\delta \equiv 2 - a = b - 2 \tag{12}$$

Then Eq. (2) becomes:

$$i_{2-\delta}(\theta) = I_{inc} / k^2 \int_0^\infty k_\delta(\alpha\theta) \; n_{\delta+2}(\alpha) \, d\alpha \tag{13}$$

where:

$$k_\delta(\alpha\theta) \equiv J_1^2(\alpha\theta) / (\alpha\theta)^\delta \tag{14}$$

Now the eigenfunctions and associated eigenvalues are then expressed in terms of the instrument configuration or design parameter δ through Eq. (14). The rate at which the eigenvalue spectrum rolls off is an indication of stability of the solution based on eigenvalue expansions, and Hirleman [14] has shown that $\delta = 0$ provides optimal inversion performance. The value $\delta = 0$ corresponds to $a = b = 2$, that is a solution on a particle area basis using log-scaled ring detectors. This result is identical to that obtained by Hirleman [12] using a condition number analysis on the linear system produced by numerical quadrature of Eq. (1).

Summary of Contributions on Inverse Scattering Algorithms

In summary, the optimal configuration for a laser diffraction instrument requires log-scaled ring detectors with the solution on a particle area basis. The number of degrees of freedom that can realistically be supported in an inversion depends on the measurement context (instantaneous noise and the size distribution), but this number can be estimated using condition number [12] or eigenvalue spectrum [14] considerations. Once the optimal order of the measurement context is determined, the corresponding optimal detector array can be configured on-line in real-time using the programmable detector array concepts developed here [10,11,15].

IV. MULTIPLE SCATTERING

The objective of this part of the research was to understand the process of multiple scattering as it might occur in future applications of intelligent particle size distribution sensors and develop inversion schemes which can operate in such an environment. A minimum requirement is that the presence of multiple scattering be diagnosed by the instrument to ensure that erroneous data not be used in control algorithms, a situation which could result in a catastrophic failure. Our objective was to surpass that and develop efficient and robust algorithms which can actually extract useful particle size information from Fraunhofer diffraction measurements in multiple scattering environments.

Multiple scattering, or the phenomenon whereby probe radiation passing through a medium undergoes more than one scattering event on its journey from the source to the detectors, significantly complicates the interpretation of the primary data obtained by optical diagnostic systems. For that reason the application of optical diagnostics in the many important systems involving optically-thick media is problematic. The objective of this component of the research was to advance the scientific knowledge base to a state where particle size distribution measurements could be made in optically thick media. The first part of such an endeavor necessarily involved understanding the *forward problem*, i.e. that of calculating the near-forward scattering properties of an optically thick medium given a known particle size distribution. The second part involved development of methods to solve the corresponding *inverse problem*, that is determination of particle size distributions from measurements of the near-froward scattering signature. Important contributions to both facets of the problem have been made and are summarized below.

Forward Problem

The phenomenon of multiple scattering significantly complicates the analysis of radiation transfer through particulate-laden media. Nonintrusive diagnostics for reacting flows depend on an understanding of light scattering and propagation, and for that reason can unfortunately not be used in the many important systems and applications involving optically thick media. Here we discuss a model developed to predict the near-forward radiative

transfer through a medium such as a dense spray where the mean interparticle spacing is greater than about three particle diameters (independent scattering regime).

The successive-orders, discrete-ordinates model developed here and discussed in detail by Hirleman [5,7,13] can calculate the near-forward scattering signature of a spatially uniform distribution of large (relative to the wavelength) particles or droplets dispersed in a medium of optical depth up to 10.0 (which corresponds to an optical extinction level of exp(-10) or 0.99995). Also, the model assumes that *independent* rather than *dependent* scattering is predominant; a condition which is met when the mean particle spacing is large compared to the particle diameters. When this independent-scattering condition is satisfied the scattering phase-function for isolated particles is used in the model, and for spherical particles or droplets the phase-function is given by Lorenz-Mie theory.

Verification of the successive-orders, discrete-ordinates multiple-scattering model [13] developed here was performed using two independent computational schemes and via experimental studies both in our lab and by other investigators. First, we developed a Monte-Carlo model for the multiple scattering forward problem which did not require the small angle assumption. Also, the discrete ordinates were not an integral part of the Monte Carlo model and therefore predictions made with the Monte Carlo code should be independent. Secondly, we worked on another independent solution to the radiative-transfer equation which involved expansion of the phase function in Legendre polynomials which provided an analytical solution for the multiple scattering case. This second method was only valid for single scattering phase functions which could be modeled acceptably well by the Legendre polynomial approximations, but these analytical scattering phase functions could be used in the successive-orders code for the test cases. Both independent numerical checks confirmed the performance of the model developed here.

In terms of experimental verification, we first compared our predictions with some limited experimental data obtained by L. Dodge of Southwest Research Institute and the Sheffield group (see [13]). The predictions of the successive-orders, discrete-ordinates model developed here compared quite well. Secondly, to support our early efforts in experimental verification a flow cell facility was developed in which standard solid calibration particles (spheres) were dispersed in liquid media. The device utilized a magnetic stirring system in an optical cell, and worked acceptably well when the particle size distributions were sufficiently narrow as was the case for the limited experiments carried out to date. These experiments also provided data which were in good agreement with predictions.

The experimental verification to this point has confirmed the validity of the model in *independent* scattering media with optical depths up to about 6 and for particles very large compared to the wavelength. It would be very interesting, however, to study the performance of the model in regimes where the size and independent scattering constraints become questionable. Further, the upper limit for validity of the model in terms of optical depth is also not clear; the numerical verification experiments indicate that calculations up to an optical depth of 10 are no problem. The results of our initial efforts to develop an experimental facility to perform these experimental studies is discussed in a following section.

<u>Inverse Problem</u>

The most interesting (and challenging) problem is that of making Fraunhofer diffraction particle size measurements in optically thick media. In this, as any inverse scattering application, a calculation method for the forward problem is necessarily an integral component of the overall system. The model discussed in the preceding section was developed with this eventual integration into a full inverse scattering instrument system in

mind. The proposed method, discussed by Hirleman [17], involves considering the multiple-scattering medium as a linear system which operates on the incident scattering signature. The scattered light signatures inside the medium are represented as vectors and are considered on a scattering order basis (as opposed to an optical or physical pathlength basis). The medium is then considered to act as a linear operator and the effect of the medium is represented as a matrix. An approach analogous to "system-identification" schemes used for dynamic systems has been proposed here to interrogate the medium and characterize on-line the scattering redistribution matrix. Numerical studies and a limited number of experiments have been performed which indicate that knowledge of the redistribution matrix will allow one to determine (through a mathematical deconvolution) the single scattering characteristics of the particle or droplet phase. Conventional inverse scattering schemes which are applicable to the single-scattering regime can then be used to recover the particle size distribution function.

In an optically *thin* particle medium the near-forward scattering signature from large particles can be deconvolved to obtain the particle size distribution function. This deconvolution or inversion process is the foundation of particle-sizing instruments based on Fraunhofer diffraction, but the ultimate accuracy of the inversion process depends directly on an ability to accurately model the light scattering processes. Unfortunately, the process of multiple scattering effectively smears out the scattering signature that would be produced by an optically thin aerosol such that scattering models which assume single scattering do not apply. Physically, one can think of a single scattering signature produced by the first layer of particles encountered by the incident laser beam, which proceeds through the medium toward the detectors (see Fig. 1). If some of this scattered light strikes another particle on the way to the detector (i.e. a multiple scattering event), then the single scattering signature will be perturbed. In effect, the scattered light is redistributed from the original scattering angle into some other generally different scattering angle. The redistribution process is caused by the multiple scattering (it does not happen to any significant extent in an optically thin aerosol), and it is necessary to be able to mathematically model the process to have any hope of recovering particle size distributions from scattering signatures which are contaminated by multiple scattering.

A conventional inverse scattering solution would take the following approach. First, the extinction and the near-forward scattering signature would be measured. Then, an initial guess of the particle size distribution and the optical depth would be made. Then, the forward or direct problem would be solved, i.e. that of predicting the scattering signature for this particle field with an assumed size distribution and optical depth. This *predicted* scattering signature would be compared to the one actually measured, and if the agreement was unacceptable an iterative process would be followed whereby the assumed size distribution and optical depth would be adjusted until the predicted and measured scattering signatures agreed to within some acceptable limits. Unfortunately, as the optical depth increases and the "smearing" effect of multiple scattering increases accordingly, the stability of the inversion process deteriorates. One symptom of the instability or ill-conditioned nature of the inversion will be that more and more size distributions will give predicted scattering signatures that are imperceptibly different than the measured scattering signature *when measurement noise is considered*, i.e. nonuniqueness. Gradually, the information in the scattering signature will be dissipated by multiple scattering, in fact in the limit of an infinite medium the scattered light would be isotropic and carry no information about the phase function of the particles in the cloud.

Our suggestion to impede this deterioration is to obtain additional more independent measurements as proposed by Hirleman [17]. In this approach, the medium is interrogated using hollow cones of light with angles corresponding to the scattered-light detector angles. The scattered light exiting at all detector angles is measured for each incident cone of light, i.e. if there are n detectors then n^2 measurements would be made as opposed to n for a

conventional single-scattering Fraunhofer system. This multi-angle interrogation effectively provides a means to measure the multiple scattering redistribution matrix on-line, which is crucial since it is the redistribution of the scattered light which renders the single scattering inversion schemes helpless. Inverting this measured redistribution matrix has the same effect as reaching into the particle medium and detecting only those photons which have been scattered exactly once - i.e. it allows one to estimate what the scattering would have been if the same particle size distribution had been in an optically thin medium.

The experimental system to allow multi-angle interrogation of the particle field is shown in Fig. 3. The key difference between Fig. 3 and conventional single scattering Fraunhofer instrument of Fig. 1 is the presence of a programmable mask in the front focal plane of the transmitter lens. The programmable mask has annular ring apertures which can be individually switched on (transmitting) or off (absorbing or opaque). A ring of light in the front focal plane of an ideal transmitter lens produces a hollow cone of light of constant angle θ passing through the spray or particle sample volume. By switching open the various rings, a sequence of hollow cones of probe radiation is created. The fraction of the incident energy in the cones which is not scattered by the medium is redirected by the transform lens to a ring on the detection plane which matches the ring in the programmable mask (assuming the focal lengths of the transmitting lens and the transform lens are equal). Light which is scattered by particles in the spray leaves at some angle different than the cone angle and ends up at another radial position (i.e. a different detector ring) on the detector plane.

Our experimental work has involved the use of optical interrogation with light from two rings. The cones of light were generated using mechanical means to spatially filter the optical energy, and the scattering signatures measured with a conventional ring detector. A portion of these results have been published by Hirleman [17] and Kenney and Hirleman [20], and the data indicate that the method is indeed feasible.

Recall from a previous section that the first use of programmable masks or spatial light modulators in Fraunhofer diffraction particle sizing systems was reported by Hirleman and Dellenback [10,11,15]. Following that work, custom spatial light modulators with semicircular ring-shaped elements as shown in Fig. 3 have been designed and fabricated, and one device has been delivered and evaluated. The contrast ratio, which was specified at 150:1, came in at about 50:1 which is marginal but probably useable for this application if lock-in amplifier schemes are implemented. The element geometry appears to be adequate, with 2 μm dimensional tolerance having been held at all measured points. A more complete discussion of this experimental work will be forthcoming [21].

Experimental Facility for Multiple Scattering Studies

The creation of a stable and well-characterized two-phase system such as a liquid droplet spray (in a gas) or a dispersion of solid or liquid particles in a liquid is very difficult. Sprays and aerosols are generally very unstable due to gravitational settling and coagulation, and despite many attempts there has been little success in using these as a reference particle field. Probably the most straightforward approach to obtain a known calibration particle system for optical instruments is to disperse glass or polystyrene calibration microspheres in a liquid using stirred- or flow-cells. As discussed above, our early efforts in experimental verification utilized such an apparatus in which standard solid calibration particles (spheres) were dispersed in liquid media. The device utilized a magnetic stirring system in an optical cell, and worked acceptably well when the particle size distributions were sufficiently narrow as was the case for the previous experiments, and some results have been reported [17,20]. However, questions concerning uniform (spatial) dispersion and centrifugal separation (by size) of the particles in broad size distributions remains. Also, the performance of the system at very high particle loadings (i.e. at or near the dependent scattering limit) is in question.

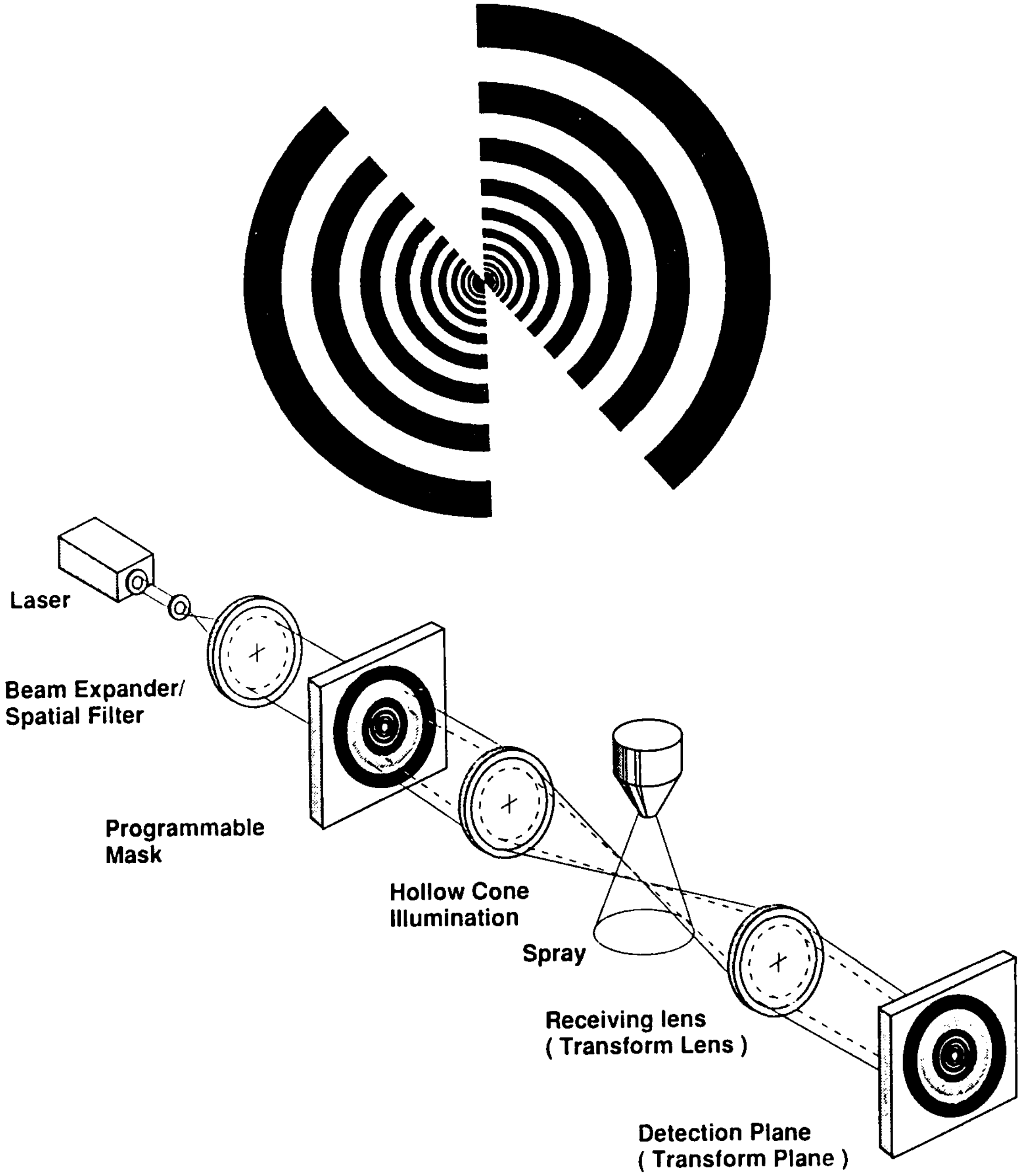

Figure 3. Schematic of optical system for multi-angle interrogation of optically thick sprays. The programmable mask on the input side transmits cylindrical shells of light which are converted to hollow cones of light by a lens. A matched programmable mask at the transform plane allows measurement of the redistribution matrix. The insert shows the actual geometry of the 34-element ring-aperture light valve designed and fabricated for this research grant.

To solve some of the problems associated with cells such as segregation and deposition we have designed and constructed a fluidized bed system to be used in the study of radiation transfer in optically thick media as shown in Fig. 4. In theory, a fluidized bed should provide a uniform dispersion of the particles and provides a very simple means by which to control the optical depth. The fluidized bed consists of the particles of interest placed in the test section between two microsieves. Water is pumped in a closed loop around the system which includes a 0.2 μm filter. When the water passes through the fluidized bed the resulting drag force on the particles will, if the velocity is higher than some critical value, overcome gravity and lift the particles. If the system is operating properly, for a given fluid velocity the bed of solid particles will expand until the overall buoyancy and drag forces are equalized. The bed should expand and contract as the flow rate is varied, and the concentration (#/cc) of particles will just depend on the height of the expanded bed and the total number of particles originally placed in the fluidizing section.

The fluidizing section in the bed developed here is 3 x 1 inch (7.62 x 2.54 cm) inner dimensions with a height of 12 inch (30.48 cm). Optical access is provided by two anti-reflection-coated quartz windows providing a 1 inch (2.54 mm) diameter clearance near the bottom of the bed and a 3 inch (7.62 cm) path length for the laser beam. (The capacity for similar windows on opposite sides of the shorter dimension is available). A Plexiglass window running the full height of the fluidizing section provides optical access to allow measurement of the bed height for the independent determination of particle concentration. With the two optical paths the system can theoretically allow a 10:1 variation in optical depth *while maintaining the same particle size distribution* by merely adjusting the flow rate. The absolute values of the optical depths are determined by the number (mass) of particles placed in the fluidizing section of the bed when it is assembled.

Our results with the fluidized bed have not been completely successful. We have been unable to stabilize the height of the bed for long periods of time, and therefore the independent measure of particle concentration has not been available. Experiments have been performed using particles in the 50-100 μm range, but our design relations indicate that the performance should be more stable for larger particle sizes. Also, we have incorporated new distributor plates, switching from wire mesh to precision photoetched plates to improve performance.

In summary, the potential benefits of a system where, for a given size distribution, the particle concentration can be varied over a large range by changing a flowrate and where the concentration can be determined independently by a simple bed-height measurement are great. However, additional work is required to achieve the desired results with the system.

Summary of Contributions on Multiple Scattering

A new successive-orders, discrete-ordinates model for predicting the near-forward scattering signature of optically thick media containing large particles in the independent scattering regime has been derived and verified. A technique for measuring particle size distributions in optically thick media has also been developed. The technique involves a novel optical interrogation scheme using pairs of ring-shaped light valves coupled with the multiple scattering model. Finally, a fluidized bed facility which has many potential advantages for associated experimental studies was developed but additional work is required to realize the full potential.

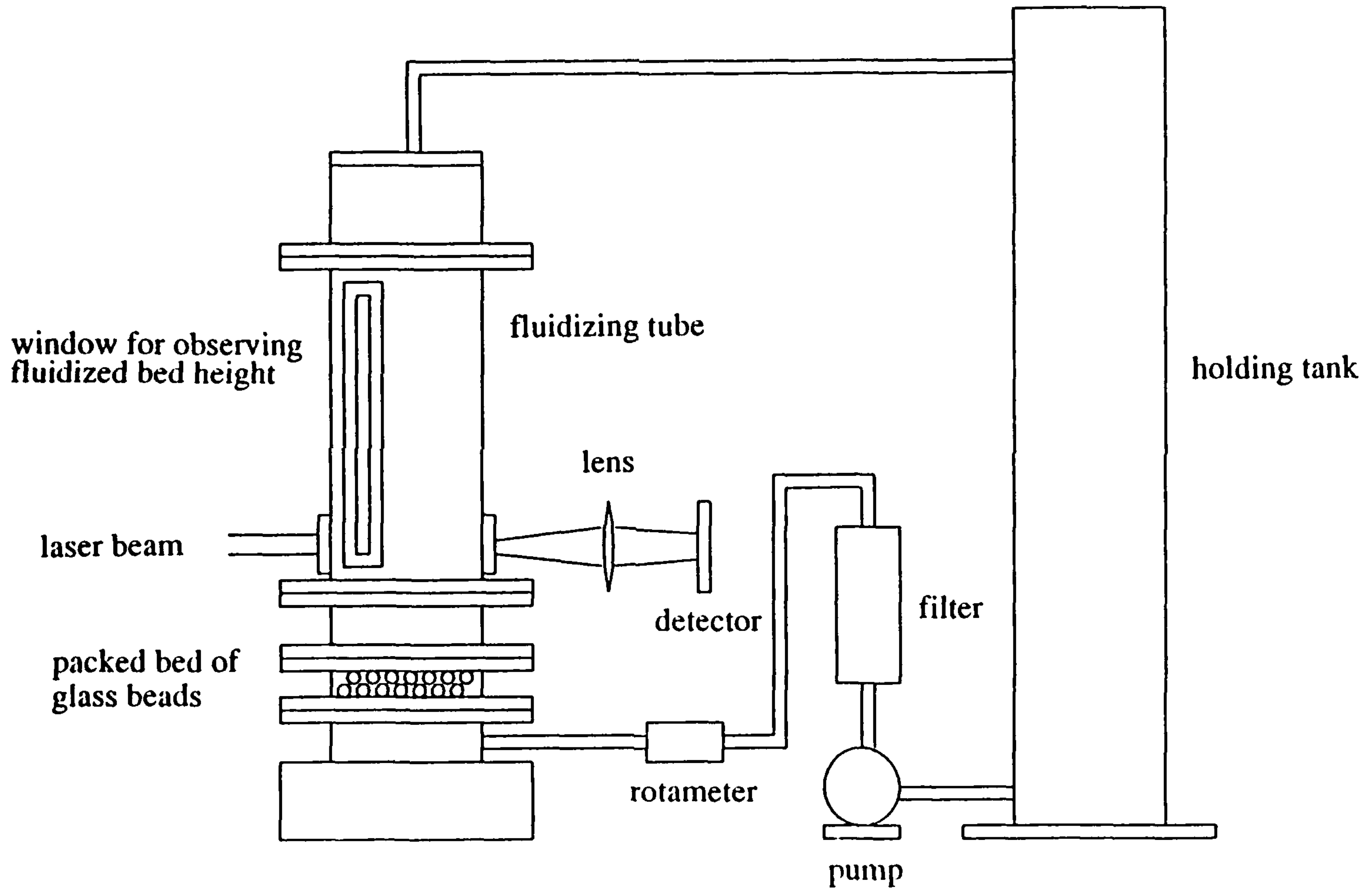

Figure 4. Schematic of Fluidized Bed Flow Apparatus.

V. PUBLICATIONS (AND REFERENCES FOR PRECEDING SECTIONS)

1. L. G. Dodge and E. D. Hirleman, "Laser Diffraction Measurements of Sprays and Reticles: Accuracy and Interpretation", Paper 85-6, Western States Section, Combustion Institute, Spring Meeting, San Antonio, TX, April 1985.

2. E. D. Hirleman and L. G. Dodge, "Performance of Laser Diffraction Drop Size Analyzers", paper 85-64, *ICLASS '85 Conference Proceedings*, The Institute of Energy, London, England, July, 1985.

3. J. H. Koo and E. D. Hirleman, "Integral Transform Inversion Methods for Laser Diffraction Particle Sizing", Paper No. 85-77, Eastern States Section, Combustion Institute, Fall Meeting, Philadelphia, PA., November, 1985.

4. J. H. Koo and E. D. Hirleman, "Comparative Study of Laser Diffraction Droplet Size Analysis Using Integral Transform Techniques: Factors Affecting the Reconstruction of Droplet Size Distributions," Paper No. 86-18, Western States Section, Combustion Institute, Spring Meeting, Banff, Alberta, CANADA, April, 1986.

5. E.D. Hirleman, "Modeling of Multiple Scattering Effects in Fraunhofer Diffraction Particle Size Analysis," Paper No. 86-17, Western States Section, Combustion Institute, Spring Meeting, Banff, Alberta, CANADA, April, 1986.

6. E. D. Hirleman, "Laser Diffraction Particle Sizing Techniques", pp. 168-175 in *Flow and Particle Diagnostics,* Vol. 58, ICALEO'86 Proceedings, Laser Institute of America, 1987.

7. E. D. Hirleman, "Modeling of Multiple Scattering Effects in Fraunhofer Diffraction Particle Size Analysis," pp. 159-176 in *Optical Particle Sizing*, G. Gouesbet and G. Grehan, eds., Plenum Publishing, London, 1987.

8. J.H. Koo, "Particle Size Analysis Using Integral Transform Techniques on Fraunhofer Diffraction Patterns," Ph.D. dissertation, Arizona State University and George Washington University, Washington, DC, 1987.

9. E. D. Hirleman, "Optimal Scaling for Fraunhofer Diffraction Particle Sizing Instruments," pp. 135-146 in *Optical Particle Sizing*, G. Gouesbet and G. Grehan, eds., Plenum Publishing, London, 1987.

10. E. D. Hirleman and P. A. Dellenback, "Faraday-effect Light Valve Arrays for Adaptive Optical Instruments", Invited Review Paper, pp. 6-10, in *Optical Methods in Flow and Particle Diagnostics*, Vol. 63, Laser Institute of America, 1988.

11. E. D. Hirleman and P. A. Dellenback, "Adaptive Fraunhofer Diffraction Particle Sizing Instrument using a Spatial Light Modulator", pp. 217-220 in *Spatial Light Modulators and Applications,* V. 8, Optical Society of America, Washington, D.C., 1988.

12. E. D. Hirleman, "Optimal Scaling for Fraunhofer Diffraction Particle Sizing Instruments," *Particle Characterization*, Vol 4, pp. 128-133, 1988.

13. E. D. Hirleman, "Modeling of Multiple Scattering Effects in Fraunhofer Diffraction Particle Size Analysis," *Particle and Particle Systems Characterization*, Vol. 5, pp. 57-65, 1988.

14. E. D. Hirleman, "Optimal Scaling of the Inverse Fraunhofer Diffraction Particle Sizing Problem: Analytic Eigenfunction Expansions," pp. 339-346 in *Proceedings of the 4th European Symposium on Particle Characterization*, Nurnberg, West Germany, April, 1989.

15. E. D. Hirleman and P. A. Dellenback, "Adaptive Fraunhofer Diffraction Particle Sizing Instrument using a Spatial Light Modulator", *Applied Optics*, Vol. 28, pp. 4870-4878, 1989.

16. J.H. Koo, M.J. Kneer, A. Chaboki, and E.D. Hirleman, "Statistical Approach to Evaluate Particle Size Inversion Algorithms," Paper 90-0151, AIAA 28th Aerospace Sciences Meeting, Reno, NV, 1990. Submitted for publication in *AIAA J. of Propulsion and Power.*

17. Hirleman, E.D., "A General Solution to the Inverse Near-Forward Scattering Particle Sizing Problem in Multiple Scattering Environments: Theory", pp. 159- 168 in *Proceedings of the 2nd International Congress on Optical Particle Sizing*, Arizona State University, Tempe, AZ, March, 1990. Submitted for publication in *Applied Optics.*

18. J. H. Koo and Hirleman, E.D., "A Synthesis of Integral Transform Techniques for Reconstruction of Particle Size Distributions from Forward-scattered Light", pp. 189-198 in *Proceedings of the 2nd International Congress on Optical Particle Sizing*, Arizona State University, Tempe, AZ, March, 1990. Submitted for publication in *Applied Optics.*

19. S. B. Kenney and E. D. Hirleman, "Particle Sizing Errors Associated with the Fraunhofer Diffraction Assumption in the Anomalous Diffraction Regime", pp. 23-26 in *Proceedings of the 2nd International Congress on Optical Particle Sizing, Post-Deadline Papers Addendum*, Arizona State University, Tempe, AZ, 1990. Also presented at the ILASS Americas '90 Conference, Hartford, CT, May, 1990.

20. S. B. Kenney and E. D. Hirleman, "A General Solution to the Inverse Near-Forward Scattering Particle Sizing Problem in Multiple Scattering Environments: Experiments", pp. 27-30 in *Proceedings of the 2nd International Congress on Optical Particle Sizing, Post-Deadline Papers Addendum*, Arizona State University, Tempe, AZ, 1990.

(The following publications are planned)

21. S. B. Kenney and E. D. Hirleman, "Measurements of Particle Size Distributions in Multiple Scattering Environments using Near-Forward Scattering", to be presented at a technical conference and published in an AIAA Journal, *Optical Engineering*, *Applied Optics*, or *Particle and Particle Systems Characterization.*

22. S. B. Kenney, "Particle Sizing in Optically Thick Media by Near-forward scattering: Theory and Experiments", M.S. Thesis, Arizona State University, expected May, 1991.

23. S. B. Kenney and E. D. Hirleman, "Edge Phenomena in Planar-diffused P on N Photodiode Detector Arrays and the Effects on Laser Diffraction Instruments", to be presented at a technical conference and published in an AIAA Journal, *Optical Engineering*, *Applied Optics*, or *Particle and Particle Systems Characterization.*

24. S. B. Kenney and E. D. Hirleman, "Fluidized Bed Facility for the Study of Multiple Scattering Phenomena in both Dependent and Independent Scattering Regimes", to be presented at a technical conference and published in an AIAA Journal, *Optical Engineering, Applied Optics*, or *Particle and Particle Systems Characterization.*

25. S. B. Kenney and E. D. Hirleman, "Scattering Models and the Inverse Laser Diffraction Particle Sizing Problem", to be presented at a technical conference and published in an AIAA Journal, *Optical Engineering, Applied Optics*, or *Particle and Particle Systems Characterization.*

VI. PROFESSIONAL PERSONNEL

Anthony M. Bruner - Undergraduate Lab Assistant and B.S. student. Has assisted the graduate students in the work, particularly regarding software and instrumentation development. Will receive the B.S. in Aerospace Engineering in Fall, 1990 and will hopefully attend graduate school.

Paul A. Dellenback - Postdoctoral research Associate. Participated in the research for 6 months at 50% effort after receiving his Ph.D. from ASU advised by another faculty member. His research contributions to this project are included in publications [10,11,15]. Prof. Dellenback accepted a position on the Faculty of Civil and Mechanical Engineering at Southern Methodist University in Dallas, TX.

E. Dan Hirleman - Professor of Mechanical and Aerospace Engineering. Principal Investigator.

Steven B. Kenney - Graduate Research Assistant and M.S. student. Using this project for his M.S. research, should receive the M.S. degree in Spring, 1991.

Joseph H. Koo - Graduate Research Assistant and Ph.D. student. Dr. Koo's Ph.D. dissertation titled "Particle Size Analysis Using Integral Transform Techniques on Fraunhofer Diffraction Patterns" was based on this research he received the Ph.D. in May, 1987 in a joint program with The George Washington University. Dr. Koo presently works in the Advanced Systems Division of the FMC Corporation.

M. V. Otugen - Postdoctoral Research Associate. Participated in the research for 3 months at 50% effort after receiving his Ph.D. advised by another faculty member. Prof. Otugen accepted a faculty position in the Mechanical Engineering Department, The Polytechnic University, Brooklyn, New York, NY in 1989.

VII. INTERACTIONS

1. E. D. Hirleman, "Calibration Techniques for Laser Diffraction and Single Particle Counting Spray Sizing Instruments", Presentation at the Fifth ASTM Symposium on Pesticide Formulations and Application Systems, Kansas City, MO, November, 1984.

2. June, 1986 visit by Prof. E. D. Hirleman to Kirtland Air Force Base, Albuquerque to visit Dr. Tim Ross and Mr. Glenn James based on preliminary contacts at the AFOSR Contractor's meeting at Stanford in June. Discussions centered on particle diagnostics requirements for research underway at Kirtland.

3. J. H. Koo and E. D. Hirleman, "Comparative Study of Integral Transform Techniques for Fraunhofer Diffraction Particle Sizing," Poster paper, First International Congress on Particle Sizing, Rouen, France, May 12-15, 1987.

4. E. D. Hirleman, "A Review of Optical Particle Sizing Techniques", Invited Seminar, Chemical Process Metrology Division, National Bureau of Standards, Washington, D.C., June, 1987. Dr Hratch Semerjian, contact.

5. E. D. Hirleman, "Droplet Size Measurement Methods", invited lecture, Project WIND Dispersion Workshop, U.S. Dept of Agriculture and U.S. Army, Kalispell, MT, August 5-7, 1987, organized by Dr. A. Stuempfle, U.S. Army, Aberdeen Proving Grounds.

6. E. D. Hirleman, "On the Inverse Fraunhofer Diffraction Particle Sizing Problem", Invited Seminar, Institut fur Thermodynamik, Universitat Stuttgart, Stuttgart, FRG, June, 1988. Prof. Frohn, contact.

7. E. D. Hirleman, "Particle Size and Velocity Diagnostics for Plasma Sprays", Invited Lecture, Propulsion Section, Motoren und Turbinen Union, Daimler-Benz, Munich, FRG, July, 1988. Dr. Klaus Rued, contact.

8. E. D. Hirleman, "The use of Near-forward Light Scattering Signatures in Determination of Particle Size Distributions", Invited Lecture, Measurements Department, Institut Franco-Allemand de Recherches de Saint-Louis (ISL), Saint-Louis, FR, April, 1989. Dr. Hans J. Pfeifer, contact.

9. E. D. Hirleman, "On the Fraunhofer Diffraction Particle Sizing Problem", Invited Lecture, Department of Information Technology, Riso National Laboratory, Roskilde, DK, April, 1989. Dr. Lars Lading, contact.

10. E. D. Hirleman, "Uncertainties Inherent in Matrix Formulations of the Fraunhofer Diffraction Particle Sizing Problem", Poster Paper, 3rd Conference of the Institute of Liquid Atomization and Spraying Systems, ILASS Americas 89, Irvine, CA, May, 1989.

VIII. PATENTS

1. E.D. Hirleman, "Programmable Detector Array Configuration for Fraunhofer Diffraction Particle Sizing Instruments", ASU Patent Disclosure, Dec. 1986. AF Invention No. 18,399 Filed for Patent Serial No. 266,952 by Hanscom Patent Prosecution Office, Department of the Air Force, 1 November 1988.

2. E.D. Hirleman, "Successive Order Model for Multiple Scattering in Fraunhofer Diffraction Particle Sizing", ASU Patent Disclosure, Dec. 1986.

3. E.D. Hirleman, "Optimal Scaling for Fraunhofer Diffraction Particle Sizing Instruments", ASU Patent Disclosure, Dec. 1986.

IX. SUMMARY OF IMPORTANT TECHNICAL CONTRIBUTIONS

1. Development, first application, and experimental characterization of the performance of programmable optical shutter arrays in laser scattering systems. References [10,11,15], and covered in Patent Application Serial No. 266,952, November, 1988. This concept can be used to obviate the beam steering problem in addition to providing for implementation of the optimal scaling laws.

2. Derivation of optimal scaling law for laser (Fraunhofer) diffraction particle sizing systems. References [12,14].

3. Synthesis of integral transform solutions to the inverse Fraunhofer diffraction particle sizing problem, including correction of literature and development of a new integral transforms. Performance comparisons on benchmark data sets. References [3,4,8,16,18].

4. Development of a new model for near-forward scattering by optically-thick particle fields. Discussed in detail in references [7,13]

5. Development of a technique for determination of particle size distributions in optically thick media. References [17,20].